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SUBJECT: The Recovery of Nuclear Materials From the One-Point Detonation Test Debris In the Metal Recovery Plant

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ABSTRACT

The results of the sand program are reported. A total of 7.8 tons of the debris containing ~~96~~ grams of plutonium and 80 grams of uranium was processed before the program was terminated in favor of the neptunium recovery program. Leaching and dissolving of the acid soluble material resulted in the removal of 95 per cent of the nuclear material in the residue. The uranium and plutonium was solvent extracted from feed solutions using pulse column and batch techniques. Based on the results presented, a recommendation is made to terminate the program.

1.0 INTRODUCTION

In the fall of 1955 the AEC requested that Oak Ridge National Laboratory assist the nuclear weapons test group in a study of the feasibility of recovering uranium and plutonium from the debris of several nuclear weapons experiments. During the tests, the nuclear materials contained in the weapons were discharged to the atmosphere and allowed to settle on 100 foot sand beds poured over an asphalt pad. The function of the sand was to retain the nuclear materials for subsequent recovery.

Sand samples taken of the beds indicated that approximately 600 grams of plutonium was retained in two test sections (areas B and C). Area D, another of the test sites surveyed, was eliminated from the recovery scheme because of wide spread contamination resulting in very small quantities of nuclear material being retained in the sand.

Based on this sampling survey, a total of 50 tons of the debris consisting of sand, rocks and wood fragments was loaded into standard 35 gal drums and shipped to the ORNL Metal Recovery Plant. The recovery program was initiated with the following objectives:

1. To obtain significant technical information from the dissolving, leaching and solvent extraction operations to complete the feasibility study.

2. To recover the nuclear material from the debris at a cost of less than $3,000 per ton of sand and at a unit plutonium recovery cost of less than $400 per gram. If the processing cost of the plutonium would exceed $175 per gram the recovery operation would be re-evaluated.

2.0 SUMMARY

A total of 50 tons of Nevada Test Pad debris, consisting of sand, rocks and wood splinters, was shipped to the Metal Recovery Plant for processing as part of the study to determine the feasibility of recovering nuclear materials from the residue. After one month of processing (March 1958) the program was terminated in favor of a program with higher AEC priority. During the program 16 per cent of the total sand, or 7.8 tons, containing 80 grams of plutonium and 80 grams of uranium was processed.

The nuclear material content of the debris processed at ORNL agreed with the results obtained from sampling the pads immediately following the weapons tests; an average of 10 to 15 per cent of the total nuclear material used for the tests was retained in the sand. A total of 400-500 grams of plutonium was estimated as the content of the 50 tons of sand collected from areas 11C and 11B. The average plutonium and uranium content of the processed material was 9.7 and 10.2 g/ton, respectively. The isotopic distribution of the uranium present in the sand was 45 and 54 per cent U-235 and U-238, respectively.

Processing of the material involved dissolving the metal drums and soluble material with nitric acid, leaching the uranium and plutonium from the sand, followed by one cycle of solvent extraction. Analysis of the residue remaining after the dissolving and leaching operation indicated that 4.8 per cent (0.5 g Pu/ton) of the plutonium was transferred to waste. Both continuous and batch processing solvent extraction methods were attempted during the program.

The total cost for the program was $72,633. The cost incurred for processing one ton of the material amounted to $9,312 with a unit plutonium cost of $955 per gram.

In view of the results presented in this report, it is recommended that the program be terminated and the remaining 42 tons of sand be transferred to the ORNL burial ground. It is felt that the cost of recovering the remaining plutonium is prohibitive based on the program objectives set forth by the AEC and the technical information gained from continued processing would be negligible. The anticipated cost for processing the remainder of the material would be $3,400 per ton.

3.0 DISTRIBUTION OF NUCLEAR MATERIALS ON DETONATION PADS

Samples of the sand of three of the circular pads were taken to determine if sufficient uranium and plutonium were retained on the pad to justify recovery. The analysis of the sand indicated that a total of 200 and 400 grams of plutonium was retained on Station B and Station C pads, respectively. Station C contained a plutonium to uranium ratio of 1.6. The third station, pad D, contained less than 6 ppm of plutonium at ground zero, therefore, chemical recovery was not attempted. Based on this survey, it was found that approximately 75 per cent of the evenly

dispersed plutonium was contained within a 25-foot radius from the zero point. Approximately 25 tons of the debris was removed from each pad and was loaded into 35-gal drums for shipment to ORNL.

The distribution of plutonium over a typical sand bed, Area C, is graphically presented in Fig. 1.

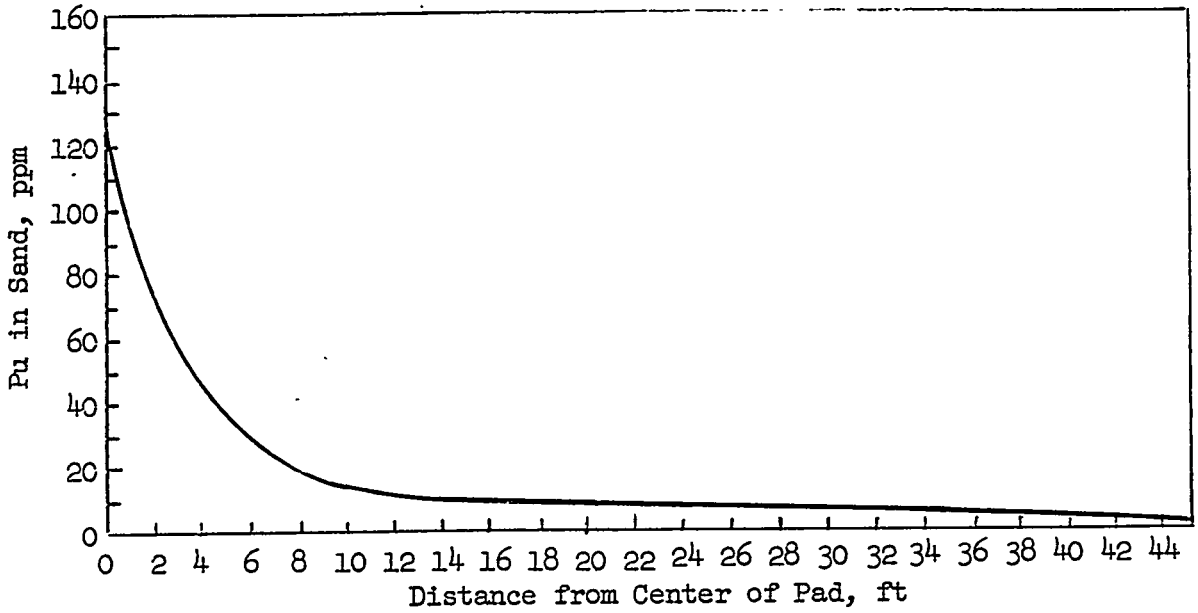

Fig. 1. Average Plutonium Distribution from Center to Outer Edge of Station C Sand Pad

4.0 DISSOLUTION

Dissolution and/or leaching of the nuclear material from the sand was accomplished in two steps which consisted of: (1) initial dissolution of the nitric acid soluble material by refluxing in the presence of excess nitric acid for 8 hours, and (2) final leaching of the residue with fresh (13 M) nitric acid. The sand used for the weapons test was of poor quality for the dissolving operation. The high carbonate content of the sand resulted in large quantities of CO_2 being released thereby causing off-gas difficulties during the dissolving operation.

4.1 Dissolution Procedure

The debris and the containers were loaded into the dissolver by way of a drum chute and crib (Fig. 2). The operational procedure used was as follows:

	Operation	Required Conditions	Purpose
1	Add H_2O (100 gal)	Ambient temperature	Dilute residual HNO_3
2	Charge drums	Tape and rubber gaskets removed	-
3	Add HNO_3 (10.0 M)	3 M HNO_3, ambient temp.	Dissolve drums
4	Heat dissolver solution	70-80°C, 1 hour	Dissolve drums
5	Heat dissolver solution	110-120°C, 8 hours (full reflux)	Dissolve nitric acid soluble material in sand. Dehydrate and coagulate silica
6	Add HNO_3 (10.0 M)	Reaction completed, 110-120°C	Increase HNO_3 strength for reuse, complete dissolution
7	Cool, sample	60°C, HNO_3	-
8	Transfer solution to feed tank using decantation	< 60°C - 5 M HNO_3	Solvent extraction feed. Remove solids from solution
9	Add HNO_3 (13.4 M) to solid residue in dissolver	80°C, 10 M HNO_3	Leach residual U and Pu
10	Transfer leach solution to surge tank	-	For reuse in the next dissolution to minimize acid consumption
11	Slurry treated sand with H_2O	-	-
12	Transfer slurry to tank farm for disposal	-	-
13	Recharge dissolver using the acid collected from step 10 as dissolution medium.		

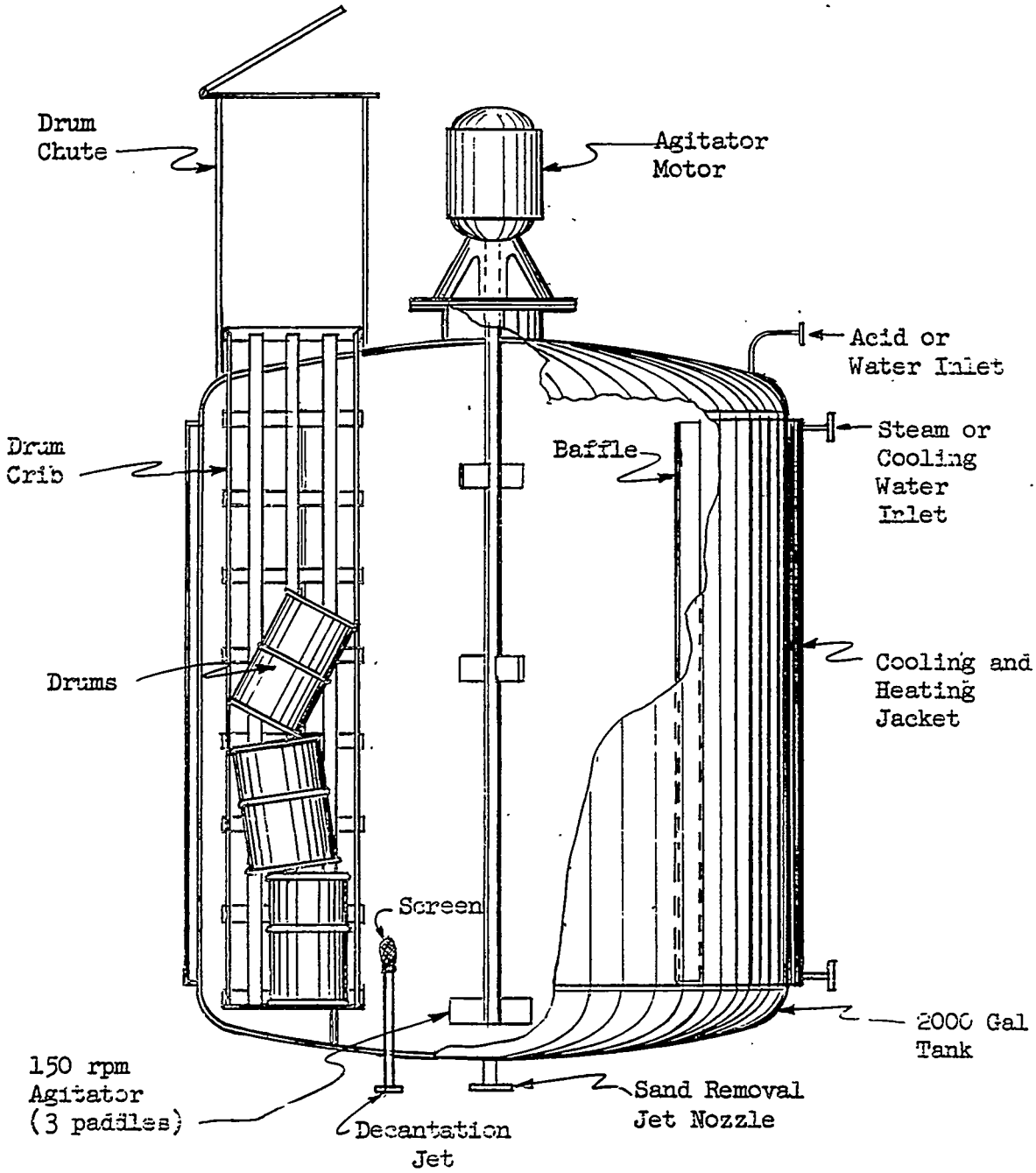

Fig. 2. Sand Dissolver - A-3.

4.2 Dissolution Results

A total of 7.8 tons of the debris was treated in the manner discussed above with a total of 76 grams of plutonium and 80 grams of uranium being leached from the material. Analysis of the sand remaining in the dissolver, after treatment, indicated 4.8 per cent (0.5 g/ton) of the plutonium could not be leached from the residues. The average plutonium content of material was 9.7 g/ton with individual batches ranging from 3.9 to 26.7 g/ton. An average of 216 gal of 13.4 M nitric acid was consumed per ton of sand for the dissolution of acid soluble material contained in the sand.

Table 4.1 presents some of the results obtained from the individual batches.

Table 4.1. Dissolving Data

Dissolving No.	No. Drums	Weight of Sand (lb)	Uranium g	Plutonium g	Grams per Ton Sand	
					Uranium	Plutonium
1	2	540	5.41	7.20	20.07	26.67
2	6	1,620	8.62	12.82	10.64	15.82
3	6	1,620	12.65	8.16	15.62	10.07
4	8	2,160	8.13	4.24	7.53	3.93
5	6	1,620	10.43	8.34	12.88	10.29
6	6	1,620	7.47	6.02	9.22	7.43
7	6	1,620	5.25	6.76	6.48	8.35
8	6	1,620	3.82	5.98	4.72	7.38
9	6	1,620	9.09	5.54	11.22	9.31
10	6	1,620	8.92	8.89	11.01	10.98
Totals or Average		15,660	79.79	75.95	10.2	9.7

4.2.1 Uranium Isotopic Distribution

The ratio of uranium to plutonium in the prepared feed was 1.05. In view of the fact that the weapons contained both natural and enriched uranium, the product solution resulting from solvent extraction was analyzed for isotopic uranium distribution. The analysis indicated the following distribution:

Isotope	Weight, per cent
U-238	54.17 ± 0.03
U-235	45.14 ± 0.04
U-234	0.48 ± 0.03
U-236	0.21 ± 0.03

5.0 SOLVENT EXTRACTION

The solvent extraction cycle consisted of a battery of two pulse columns. The uranium and plutonium were extracted from the feed solution in the first contactor using 15 per cent TBP as the extractant. In the second column the uranium and plutonium were stripped from the organic phase with 0.5 M HNO_3 and 0.008 M ferrous sulphamate. The product solution resulting from the solvent extraction cycle was continuously volume reduced in a pot type evaporator. The recovery of plutonium and uranium using the flowsheet presented in Fig. 3 follows established technology therefore a discussion on the flowsheet will be eliminated.

The plutonium recovery efficiency during the equilibrium portion of the solvent extraction cycle was 89 per cent, however, the entrainment of organic silicious material to the product evaporator and poor strip column phase separation gave difficulty in the product concentration step. Small quantities of organic, containing most of the plutonium, would collect on the top of the boiled down product. The use of 0.1 M Na_2CO_3 as a uranium and plutonium stripping reagent was attempted with only partial success.

It is felt that with an efficient method for filtering the feed solution and with good phase separation of the aqueous product solution, the recovery scheme could be vastly improved.

5.1 Batch Treatment

In view of the difficulties encountered using continuous solvent extraction, the last batch of virgin feed was treated by a tank batch extraction technique. Four successive extractions were made into 265 gallons of 15 per cent TBP in Amsco. A total of 14 grams of plutonium was recovered from the 18 grams of plutonium (75.2 per cent recovery). Stripping of the extracted plutonium was accomplished by contacting the solvent with 0.01 M

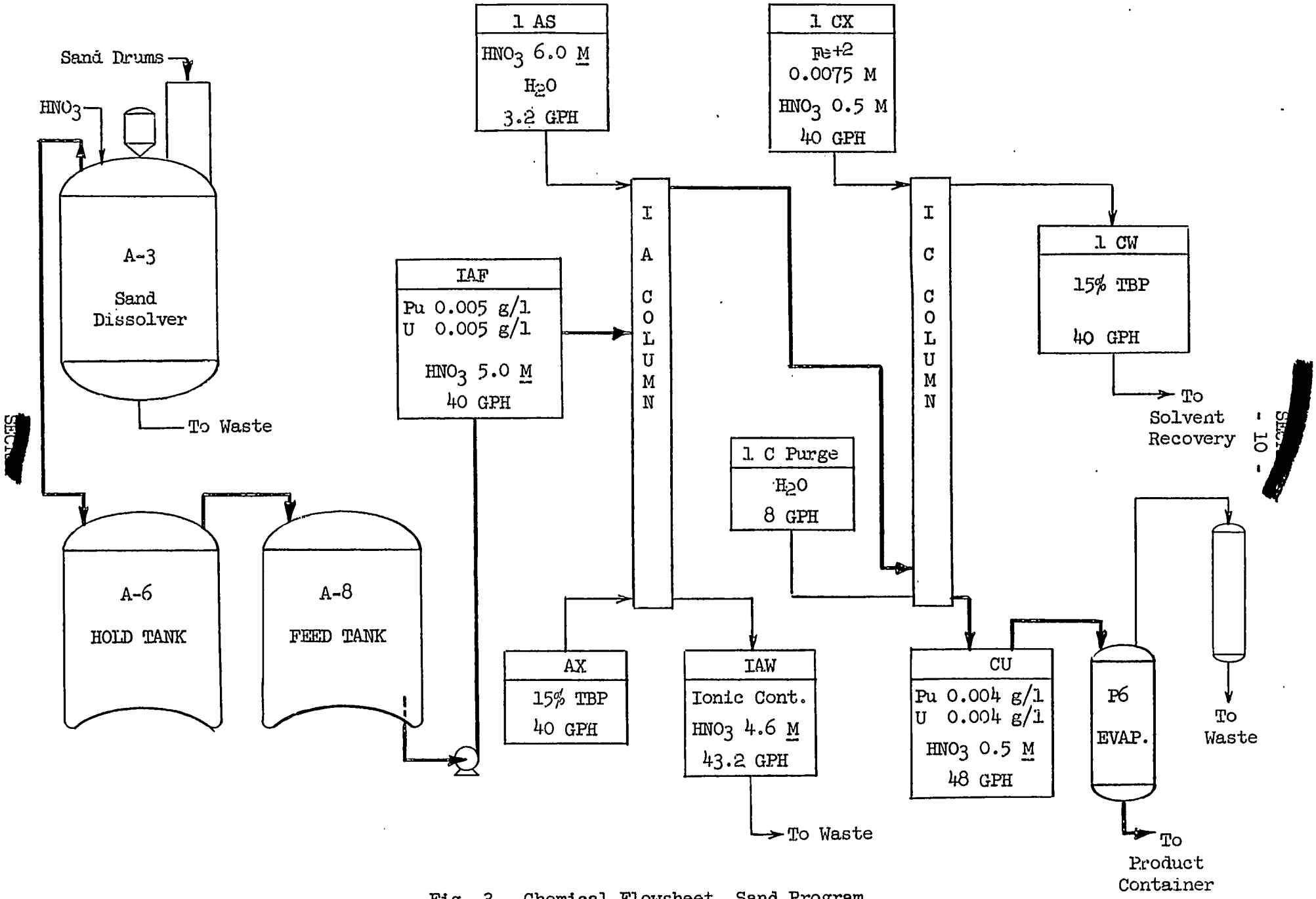

Fig. 3. Chemical Flowsheet, Sand Program

ferrous sulphamate solution. The organic/aqueous ratio for all of the contacting operations was 0.53. The major loss occurred during the stripping phase.

6.0 PROGRAM COST ANALYSIS

The total operating cost of the recovery program amounted to $72,633. The sand was processed at a unit cost of $9,312 per ton of sand and a corresponding plutonium recovery cost of $955 per gram. The distribution of the costs accumulated during the program was:

	Process Cost	
	$/ton sand	$/gram Pu
Process equipment modifications[1]	187	19.30
Pre-operating costs[2]	1,135	117.00
Operating costs	6,100	625.00
Post operating costs[3]	1,890	194.00
TOTALS	9,312	955.30

1. Includes alterations for drum handling, dissolver revisions and process piping costs.

2. Prorated from operating costs incurred during equipment modifications and flowsheet testing.

3. Prorated from operating costs incurred during equipment decontamination and the dismantling of equipment.

The anticipated costs for recovering the remainder of the nuclear material from the sand would be approximately $3,400 per ton.

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